LA-UR- 02-7615

Title: **Fission Barrier Properties, Resonance Fluctuations and Isomer Fission Cross-Sections**

Author(s): J. Eric Lynn, T-16
Anna C. Hayes, T-6

° Los Alamos
NATIONAL LABORATORY

73

FISSION BARRIER PROPERTIES, RESONANCE FLUCTUATIONS AND ISOMER FISSION CROSS-SECTIONS

J. ERIC LYNN AND ANNA C. HAYES

Group T-16, Los Alamos National Laboratory, New Mexico, USA, 87545

Although the main picture of fission barrier physics was established some time ago many of the details still have to be settled. Consequently, the application to evaluation of cross-sections of unmeasurable or exotic nuclides and their excited states is still in its early stages. In this paper I consider some of these details and explore the possibility of quantitative estimation of fission cross-sections.

1. Introduction

Our present understanding of the early stages of the fission process (i.e. the passage across the barrier) is largely based on the intensive experimental and theoretical work of the 1960s and 70s (for a review, see [1]). This gave us an appreciation of the role of nucleon shells in the deformed nucleus and their effect on both the topography of the potential energy surface and on the inertial parameter. Of course earlier work, especially that of A. Bohr [2] on the role of saddle point channels with special properties such as that of carrying a good K quantum number as well as total angular momentum and parity, remained as very valuable insights into the fission process and could be carried over into the new picture of the multi-humped barrier. All this information and understanding offers a good prospect of estimating with some confidence, at least in the lower neutron energies, the cross-sections of very heavy exotic nuclides in which fission is a significant reaction mode. In recent years the demand for such cross-section data for applications ranging from nuclear astrophysics to nuclear criticality safety has become considerable. In this paper we review the current work at Los Alamos on models and calculations of neutron-induced fission cross-sections and those of competing cross-sections.

1. Intermediate structure

A major feature in the topography of the fission barrier of the actinides is the existence of a secondary well between inner and outer barrier peaks [3]. This gives rise to shape meta-stable states (class-II states) that are very complicated at the excitation energies for which fission becomes a significant competing reaction in compound nucleus decay from the primary well. The statistical model for dealing with this feature in the overall barrier postulates a transmission coefficient T_A for the deforming nucleus to cross the inner peak, A, whereupon the system equilibrates into the complicated class-II states of the secondary well. The

probability of these states to decay by fission rather than to return to the primary well is $T_B / (T_A + T_B)$ where T_B is the transmission coefficient for making the transition across the outer peak B. The overall transmission coefficient for the class-I states of the primary well to decay by fission is therefore $T_F = T_A\ T_B / (T_A + T_B)$. This is the transmission coefficient for fission that would be used in standard Hauser-Feshbach theory.

Near and below the barrier peaks the class-II states manifest themselves as intermediate structure in the fission cross-section. This has very important implications for estimating fission cross-sections. Because the fission strength is clustered into intermediate resonances, the average fission cross-section over many intermediate resonances is lower than the value given by the statistical model. This reduction effect can be modelled [4] by a uniform "picket-fence" of class-II states with equal spacing D_{II} with the same values of coupling width $\Gamma_{II'}$

$= T_A\ D_{II} / 2\pi$ and the fission width $\Gamma_{II(F)} = T_B\ D_{II} / 2\pi$. Below the barrier peaks this reduction can be as much as an order of magnitude.

There are also important effects from quantum chaos in both the class-II states and the class-I states. In the class-II states the factors to be considered are:
a) the Porter-Thomas fluctuations of the partial fission widths through the saddle-point channels of the outer peak,
b) the Porter-Thomas fluctuations of the coupling width to the class-I states through the inner barrier,
c) the Wigner-type fluctuations of the class-II level spacings.

In the class-I states we must consider:
a) the Porter-Thomas fluctuations of the squared coupling matrix elements of the individual class-I states to the closest class-II states,
b) the Porter-Thomas fluctuations of reduced neutron widths for elastic and inelastic scattering,
c) class-I level spacing fluctuations; when intermediate structure is sufficiently sharp to be describable by perturbation theory the fluctuation of the position of the class-I levels close to a class-II level is particularly important.

An analytical approach to the problem of averaging over all these fluctuations seems intractable. We have therefore resorted to a Monte Carlo approach, in which individual values of the parameters listed above are selected, using pseudo-random numbers, from the appropriate distribution functions with specified mean values. The eigenvalue problem is solved to give R-matrix resonance parameters across the whole range of an intermediate resonance. From these the cross-section is calculated and averaged. A large number of such calculations for the specified mean values yields an ensemble of σ_F values from which the mean value of the cross-section and the variance, skew and maximum likelihood estimator of the local cross-section averaged over individual class-II states are obtained.

The averaging factor S is defined as the ratio of the average cross-section $<\sigma_F>$ calculated by this procedure to the cross-section calculated from the uniform

picket-fence model, $<\sigma_F> = S<\sigma_{UPF}>$. Some calculated results are shown in Fig. 1 as contours of S in the plane of inner and outer barrier transmission coefficients. The particular case shown is for neutron energy 10keV, mean class-I level spacing of 1eV and mean class-II level spacing of 50eV. These and other parameters are similar to those encountered in actinide nuclei. The low values of S over large parts of the plane demonstrate the importance of taking this factor into account in analyzing and estimating fission cross-section data.
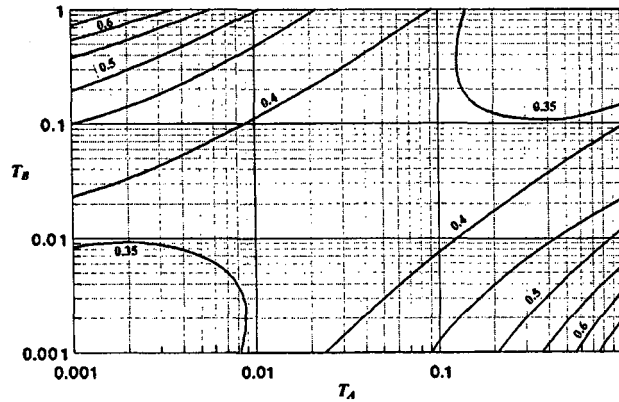
Figure 1. Contours of the averaging factor S as a function of T_A and T_B.

Towards the bottom left-hand corner of the plane shown in Fig.1 (low T_A and T_B values) the distribution of σ_F values becomes increasingly extreme. In Figure 2 we show the ratio of the variance of σ_F to the squared mean $<\sigma_F>^2$ as a function of T_B for a few T_A values. A ratio of unity is equivalent to an exponential

Figure 2. Variance of fission cross-section. T_A : 1.0——, 0.1——+——, 0.01——*——

distribution and a ratio of two to a Porter-Thomas distribution. The reason for much higher values of $\mathrm{var}\,\sigma_F / <\sigm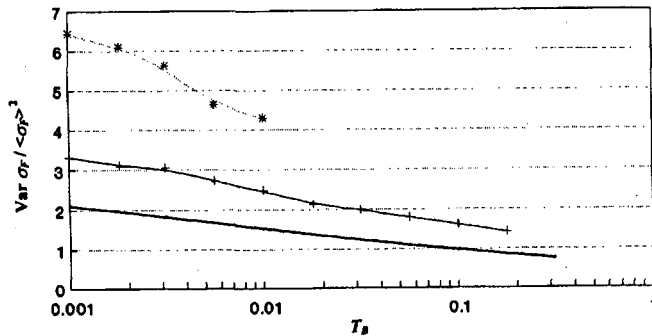a_F>^2$ is that the class-II levels become very narrow and no longer encompass many class-I levels within a Lorentzian envelope. Perturbation theory becomes applicable and the magnitude of the individual value of σ_F depends on the degree of accidental degeneracy between the class-II levels and the nearest class-I levels. For much lower values of the transmission coefficients than those shown in Figure 1 we expect the averaging factor S as defined above to fall to even lower values because of this perturbation regime.

3. Transmission spectra of even compound nuclei

The method for calculating the transmission coefficients T_A, T_B are based on A. Bohr's saddle-point channel postulate [2] and the Hill-Wheeler penetration factor formula [5] for quantal tunnelling of a barrier with the form of an inverted harmonic oscillator. At low neutron energies the details of the transition state spectrum are paramount in governing the fission cross-section, whereas at higher energies statistical level density models are sufficient to describe the cross-section, the number of fully open saddle-point channels by then being very large.

The modelling of the transition state spectrum of an even fissioning nucleus is based on the well-known collective structure of actinide nuclei with normal deformation. Thus we postulate a "ground" state (at the saddle) carrying a rotational band $J^\pi = 0^+, 2^+, 4^+, 6^+,$; a mass-asymmetry vibration with spin-projection along the symmetry axis $K^\pi = 0^-$, and associated rotational band $J^\pi = 1^-$,3^-, 5^-, ; a bending vibration with $K^\pi = 1^-$, and associated rotational band $J^\pi = 1^-$,2^-, 3^-, ; a gamma vibration (or rotation - see below) with $K^\pi = 2^+$, and associated rotational band $J^\pi = 2^+, 3^+, 4^+,$; and combinations of these.

The energies of these various state are dictated by the potential energy surface in deformation space at the relevant barrier. The moments of inertia for the rotational bands are assumed to follow the rigid-body form and are extrapolated from the observed values of rotational bands at normal deformation. At the inner barrier the bending vibration is assumed to be at 0.8MeV and at the outer barrier to be at 0.6MeV.

The mass asymmetry and gamma modes require special consideration. Calculations [6] using methods based on Strutinsky's liquid-drop-plus-shell-correction theory demonstrate that while the inner saddle point has a mass symmetric form the outer saddle is stabilized at a mass-asymmetric shape for the main actinides, and this feature is well supported by experiment in the thorium isotopes. Therefore, the mass asymmetry vibration is assumed to be at 0.7MeV at the inner barrier but to have a low value (0.1MeV) at the outer barrier because of the low tunnelling frequency between the mirror-image saddle point shapes

through the mass symmetric hill. On the other hand, theoretical calculations indicate that the inner-saddle is axially asymmetric [7]. The energy difference between the axially symmetric and asymmetric shapes is least in the uranium isotopes, so it is necessary to explore a range of models for this feature in calculating fission cross-sections for these. In Model 1 we assume normal axial symmetry with the transition state energy as 0.8MeV for both barriers. In Model 2 we assume the axial asymmetry parameter γ to be 11°; this gives a gamma rotation with spin $J^{\pi} = 2^{+}$ and energy about 250keV and a second member of the gamma rotational band with spin $J^{\pi} = 4^{+}$ and energy about 1MeV. In Model 3 the degree of axial asymmetry is maximized at $\gamma = 30°$, and within the energy gap there are at least three members of the gamma rotational band with spin $J^{\pi} = 2^{+}$, energy ≈60keV, $J^{\pi} = 4^{+}$, energy ≈200keV and $J^{\pi} = 6^{+}$, energy ≈400keV. In Model 4 we assume an axially asymmetric form that is soft to axial deformation with a basic gamma vibration energy of 0.25MeV.

As in normally deformed nuclei we expect that these collective states and their low energy combinations will lie within the energy gap due to pairing forces. Above this energy gap a large number of 2 quasi-particle states will arise. These will act as heads for vibration-rotation bands. In this region we assume therefore that the transition state spectrum can be described by a statistical level density form. The transition state density above the energy gap $E_{G(A,B)}$ is assumed to have the simple temperature form:

$$\rho_{A,B}(I^{\pi}) = C_{A,B}(2J+1)\exp[-(J+1/2)^2/2\sigma_{A,B}{}^2]\exp(U/\Theta_{A,B}) \qquad (1)$$

where U is the energy above the barrier. Because these denser states above the energy gap will affect significantly the cross-section at power energies it is necessary to take them into account through integrals convoluting the level density with the Hill-Wheeler formula.

4. Application to neutron-induced fission of odd-mass uranium targets

The discrete transition state spectra within the energy gap are essentially sufficient to allow the determination of the barrier heights of the even mass uranium nuclides from particle transfer induced fission reactions, such as (d,pf) and (t,pf). With Model 1 the inner barrier height is found to be considerably lower than the outer barrier, but with models 2 to 3 the two barriers are essentially equal and approximately 1 MeV below the neutron separation energy of ^{234}U and ^{236}U. The difference is closer to 0.5 MeV for ^{238}U. We expect therefore that the top of the energy gaps at the barriers will appear a few hundred keV or less above the neutron separation energy of ^{234}U and ^{236}U. The residual states for inelastic neutron scattering out of these compound nuclei appear to be rather completely

known (at least for the residual nucleus ^{235}U, which can also be used as a guide for completing an assessment of the likely level scheme for ^{233}U). Therefore, from the fission cross-sections and calculated inelastic neutron competition of these target nuclides we can fit the density of transition states above the energy gaps.

In fitting the data for the target nucleus ^{235}U the spin dispersion coefficient is assumed to have the value 6. The energy gaps and transition density coefficients are given in Table 1. The energy gap for the inner barrier seems very high in Model 1, and for this reason we feel that this model is less viablel than the others.

Table 1. Deduced level density parameters at barriers and for residual nucleus. All energies are in MeV or reciprocal MeV units.

| | Model 1 | | Model 2 | | Model 3 | | Model 4 | |
	A	B	A	B	A	B	A	B
Barrier	5.2	5.7	5.53	5.53	5.53	5.53	5.53	5.53
$E_{G(A,B)}$	1.65	1.03	1.25	1.15	1.32	1.22	1.1	1.15
$C_{A,B}$	0.20	0.05	0.34	0.07	0.34	0.07	0.34	0.07
$\Theta_{A,B}$	0.42	0.42	0.47	0.47	0.46	0.46	0.47	0.47
C_R	0.19		0.21		0.22		0.19	
Θ_R	0.54		0.57		0.50		0.55	

Above about 1.2 MeV neutron energy we must describe the level density of the residual nucleus by a simple statistical formula. We use the temperature form of Eq.1. Extrapolating the barrier level density parameters of Table 1 into this energy region we find the residual nucleus parameters listed in the last 2 lines of the Table.

The transition state spectra and barrier and level density parameters of Table 1 enable us to make good fits to the fission cross-section of ^{235}U. With inner and outer barrier heights of 5.83MeV for ^{234}U we can also fit the cross-section of ^{233}U very well using the other parameters of Table 1.

5. The fission cross-section of the isomer of ^{235}U

These models of the transition spectra can be extended to other nuclides for which no or little experimental data on the fission cross-sections are available. A particularly interesting case is that of the 77eV isomer of ^{235}U with half-life 25m. Measurements of the thermal neutron cross-section have shown that this is about twice as large as the cross-section of the ground state. This has led to the speculation that the fast neutron fission cross-section will also be higher causing the calculated fission yield in a very intense neutron flux to be increased, and conversely that the capture yield (to the product ^{236}U) will be lowered. The

argument is not strong because of the highly stochastic nature of thermal neutron cross-sections. We have therefore used our models to calculate the isomer cross-section over the energy region from 1keV to 2MeV to test this speculation.

At neutron energies up to a few tens of keV s-wave neutron absorption to form the compound nucleus is dominant. Above that, p-wave neutron absorption becomes dominant and remains so until well above 0.5MeV. It follows that only a few saddle-point channels out of the total transition state spectrum are important for the fission cross-section in the lower part of the neutron energy range. Because the target spins I^π of the ground state and isomer of ^{235}U are very different ($7/2^-$ and $1/2^+$ respectively) the important saddle-point channels for the relevant compound nucleus spins J^π are quite different for the two states. These are shown in Table 2; only the lowest states in the transition state spectra are listed.

Table 2. Saddle-point channels for the ground state and isomer of ^{235}U (Model 3).

I^π	l	J^π	CN weight	Trans.state	$V_A(J^\pi) - S_n$	$V_B(J^\pi) - S_n$
Ground:						
$7/2^-$	0	3^-	7/16	mass asym.vib.	0.3	0.9
		4^-	9/16	bend.vib.+rot.	0.2	0.5
	1	2^+	5/16	rotation	1.0	1.0
		3^+	7/16	γ-vib.+rot.	0.9	0.1
		4^+	9/16	rotation	0.9	0.9
		5^+	11/16	γ-vib.+rot.	0.9	0.0
Isomer:						
$1/2^+$	0	0^+	1/4	zero excn.	1.0	1.0
		1^+	3/4	bend.vib.+m.a.	-0.5	0.3
	1	0^-	1/4	2q.p.(> en.gap)	-ve	-ve
		1^-	3/2	bend.vib.	0.3	0.9
		2^-	5/4	bend.vib.+ rot.	0.2	0.5

Table 2 shows that s-wave neutron-induced fission will be considerably suppressed and to a much smaller extent p-wave induced fission will be reduced as well. Detailed calculations verify this expectation. In Figure 3 we show the experimental fission cross-section data of ^{235}U and the Model 3 calculated cross-section for both the ground state and the isomer. Contrary to the expectation from the high value of the thermal neutron cross-section, the isomer cross-section is only about half the value of the ground state cross-section at low neutron energies and the two cross-sections do not attain near-equality until about 0.5MeV neutron energy. The capture cross-section of the isomer is correspondingly enhanced.

6. Neutron-induced fission of even U nuclides

Most of the longer lived nuclides have neutron separation energies below the barrier. Therefore, information on their barrier height parameters can be determined from the neutron-induced fission cross-section. Transition states are taken from calculations [8,6] of neutron single-particle orbitals at deformations
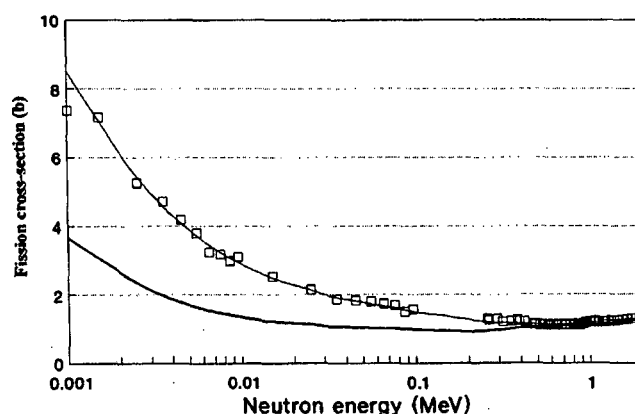

Figure 3. The fission cross-section of ^{235}U and calculated cross-section for the ground state (light line) and the isomer (bold line).

corresponding to the inner and outer barriers; to these are added vibration-rotation bands as prescribed for the even compound nuclei (Section 3). We find that the single-particle level density has to be about double that of normal odd-neutron nuclei in order to reproduce the magnitude of the fission cross-sections. The determined barrier heights are $V_A = 5.45$ MeV, $V_B = 5.65$ MeV for the compound nucleus ^{235}U, $V_A = 5.9$ MeV, $V_B = 5.7$ MeV for the compound nucleus ^{237}U, and $V_A = 5.95$ MeV, $V_B = 5.85$ MeV for the compound nucleus ^{239}U. Above the energy gap of the target nucleus the density of states for inelastic scattering to the residual nucleus are found to be consistent with the parameters $C_R \approx 0.1$-0.15MeV^{-1} (considerably lower than C_A for the corresponding even fissioning nucleus), $\Theta_R \approx 0.5$MeV.

The fission cross-section of ^{232}U is of considerable interest, especially in nuclear criticality safety. There are few experimental measurements and these are incomplete and inconsistent. The downward trend of barrier heights with decreasing mass number in the odd uranium nuclei that we observe above suggests that $V_A = 5.55$ MeV, $V_B = 5.55$ MeV might be a reasonable choice of barriers for ^{233}U. This gives the theoretical curve shown in Fig.4, where it is

compared with the three available sets of experimental data [9]. The theory helps eliminate one experimental set and agrees quite well with the others.

7. Fission cross-sections of other actinide isomers

^{235}U is unusual in having an isomer in that it is an even-odd nuclide. Most isomers occur in odd-odd nuclides because the lowest 2quasi-particle state can occur quite close in energy with the spins of the odd particles either aligned or anti-parallel, giving, often, a very large K and spin difference. In ^{235}U, the long
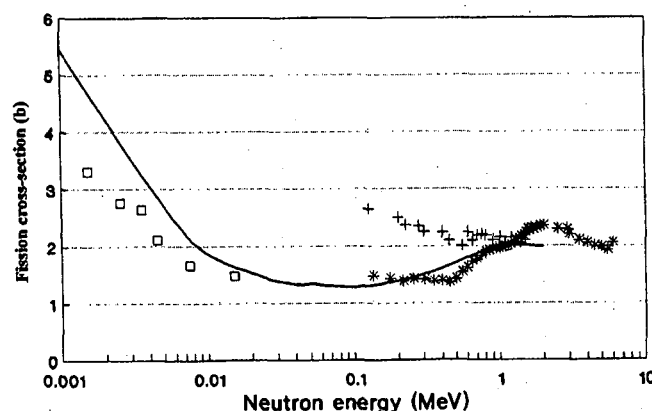

Figure 4. Fission cross-section of ^{232}U.

half-life of the isomer appears to be due primarily to the extremely small energy difference between it and the ground state.

We have not studied the fission cross-sections of the odd-odd actinides. However, a general remark is in order. The neutron separation energy of the compound nucleus is generally significantly higher than the fission barrier. The transition state spectrum of the compound nucleus will be quite dense, with vibration-rotation bands built on single proton quasi-particle states. It follows that there will be many more saddle-point channels available at low neutron energies for compound nucleus spins derived from a high-spin target nucleus than from a low-spin target. The high-spin target can thus be expected to have a considerably higher fission cross-section than its low spin counterpart.

References

1. S.Bjornholm and J.E.Lynn, *Rev.Mod.Phys.* **52**, 725 (1980)
2. A.Bohr, *PeacefulUses of Atomic Energy* **2**, 220 (United Nations, 1955)
3. V.M.Strutinsky, *Nucl.Phys.A*, **95**, 420 (1967)
4. J.E.Lynn and B.B.Back, *J.Phys.A* **7**, 395 (1974)

5. D.L.Hill and J.A.Wheeler, *Phys.Rev.* **89**, 1102 (1953)
6. P.Moller and J.R.Nix, *Phys.&Chem.of Fission* **1**, 103 (1974)
7. S.E.Larsson and G.Leander, *Phys.&Chem.of Fission* **1**, 177 (1974)
8. S.G.Nilsson *et al., Nucl.Phys.*A **131**, 1 (1969)
9. B.I.Fursov *et al.At.En.***61**, 383 (1986)